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Fallout β-activity in precipitation

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With 2 figures in the text

Introduction

For several years the fallout over Sweden from nuclear weapon tests has been systematically measured at this laboratory. This paper is intended to review some of the information obtained in these studies in the years 1953–57. It contains results of β-measurements on collected samples of rainwater and snow. It is planned that other aspects of this work will be described in a coming paper.

Sampling procedures

The measurements reported have been made on samples of precipitation collected at a place close to Stockholm. The collection device consists of a stainless steel funnel with a diameter of 2 m and a cone angle of 120°. The funnel is fitted with a cylindrical edge of 1 dm height, also of stainless steel. The upper funnel surface is situated about 2 m above ground level to prevent dust from the ground to contribute to the collected samples. The funnel itself forms the roof of a small wooden house in which is placed a heat source and a collection vessel. The heat source is used for the melting of snow and is turned on if necessary at the end of the precipitation interval. It has been found that 1 kW of heating power is sufficient for most practical purposes. Up to January 1956 the samples were collected in glass or polythene bottles with a volume of about 60 l. Samples were taken in this way during each rain or snow fall. The collected water was transported to the laboratory where preparation of the sample was made. This preparation consisted of evaporation of the water to dryness in a number of 3 l porcelain dishes. The residue was carefully collected and ashed at a temperature of between 300° and 400°. After this treatment the sample was placed on a source dish of aluminum 30 mm in diameter. The above procedure was in January 1956 replaced by another method introduced by Edvarson [1]. According to this method the water is led through a polythene collecting unit containing paper pulp, filter paper, anion exchanger (Dowex 2) and cation exchanger (Dowex 50). This collecting unit is small in volume and esaily transportable by mail. The preparation of these samples consisted of separate ashing of the paper and ion exchanger fractions. After ashing, the sample was mounted as before on an aluminum source dish. In all preparations mentioned above the sample was fixed by a solution of Zapon laquer in amylacetate.

Measurements of fallout β-activity

All collected samples were measured for their total β-activity as a function of time. This was done with an end window GM-counter [2] mounted in a 5 cm thick lead shield to reduce the background. The detector unit was combined with a sample changer with a capacity of 24 samples. Each sample was measured for about 1 hour. For every four samples the back-ground was measured and the calibration checked with a standard source. This standard source consisted of a uranium glass piece contained in an aluminum source dish of the same type as those used for the fallout samples. The efficiency of the counting arrangement has been calibrated using reactor produced fission products and found to be about 10 %.

The decay of each sample was followed for about 8 weeks although some strong samples have been measured for over a year.

Results and discussion

The fallout rate in mc/km² and week for the period from April 1953 to January 1958 is given in Fig. 1.

An inspection of the individual decay curves shows that most samples containing debris younger than about 100 days have given decay curves that fits rather well with the $t^{-1.2}$ curve during the first few months of measurement. Later a deviation in form of slower decay than expected is often found. This indicates an admixture of older debris, of one or more origins. An age determination of the younger component can be made from the first part of the decay curve. When the samples contained only relatively old (more than about 100 days) activities or the young component was weak compared to the older, the effect of admixture often makes it impossible

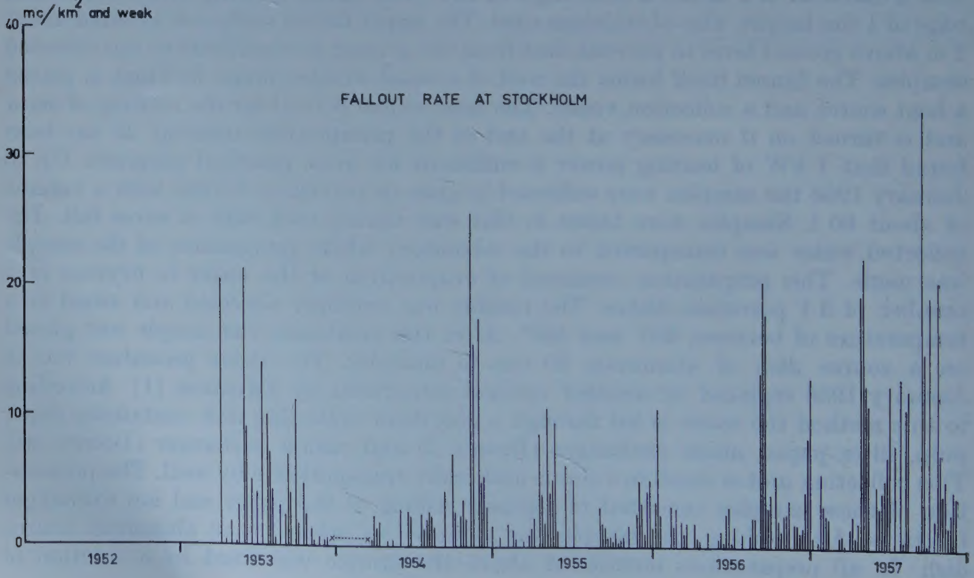

Fig. 1.

Table 1

Period of high fallout rate	Test date	Test country
April–May –53	March–May –53	USA
Sept–Oct –53	Aug –53	USSR
Nov –54	Aug–Oct –54	USSR
April–May –55	March–April –55	USA
Dec –55 –Jan –56	Sept–Nov –55	USSR
Sept –56	Aug–Sept –56	USSR
Dec –56 –Jan –57	Nov –56	USSR
May –57	March–April –57	USSR
July–Aug –57	June–July –57	USA
Sept–Oct –57	Aug–Sept –57	USSR

to identify any pure component in the decay curve although some sort of "mean" age can be estimated and used for the extrapolation beyond the period of measurements.

The dominating peaks in Fig. 1 can be fairly well identified as they represent rather young samples of high activity. Each period of high fallout rate can thus be attributed to a known test series, see table 1. As is found from the table, the high fallout rates occur within 1–3 months after the test dates.

Fig. 2 shows the accumulated fallout deposit up to July 1957. This has been calculated as the sum of the individual fallout sample decay curves. These decay curves have been measured until the activity has decreased to a small percentage of the total deposit and then extrapolated with the help of the $t^{-1.2}$ decay law.

Accuracy of measurements and age determinations

With the sampling area used, a normal rainfall as a rule has given enough activity to make the statistical error less than 1 % in the routine measurements. This is probably negligible compared to the errors due to self-absorption in the sample and to variation of efficiency with β-energy. As the samples as a rule have been of about the same thickness it was considered justified to use a standard correction for self-absorption.

The accuracy of the age-determinations is influenced by other factors than the measuring errors. Thus the $t^{-1.2}$ law is only an approximation and the deviations are of the order of magnitude ± 20 % [3]. In young samples the activity of neutron induced actinides (Np^{239} and U^{237}) is often quite appreciable, which makes the deviation from the $t^{-1.2}$ law still larger. It is estimated that the loss of volatile compounds (iodine, noble gases) during ashing of the samples is less than 10 % of the total activity. Further it has been found [4] that large particles of high activity have a composition different from mixed fission products, probably due to chemical fractionation during the particle formation period in the first minutes after the explosion. Sometimes a large part of the activity is carried by such particles. The occurrence of mixed activities of different ages complicates the age-determination as has been discussed above.

The only way to estimate the composite error due to all these contributions is to compare the calculated age with known test dates. This can be done when a single

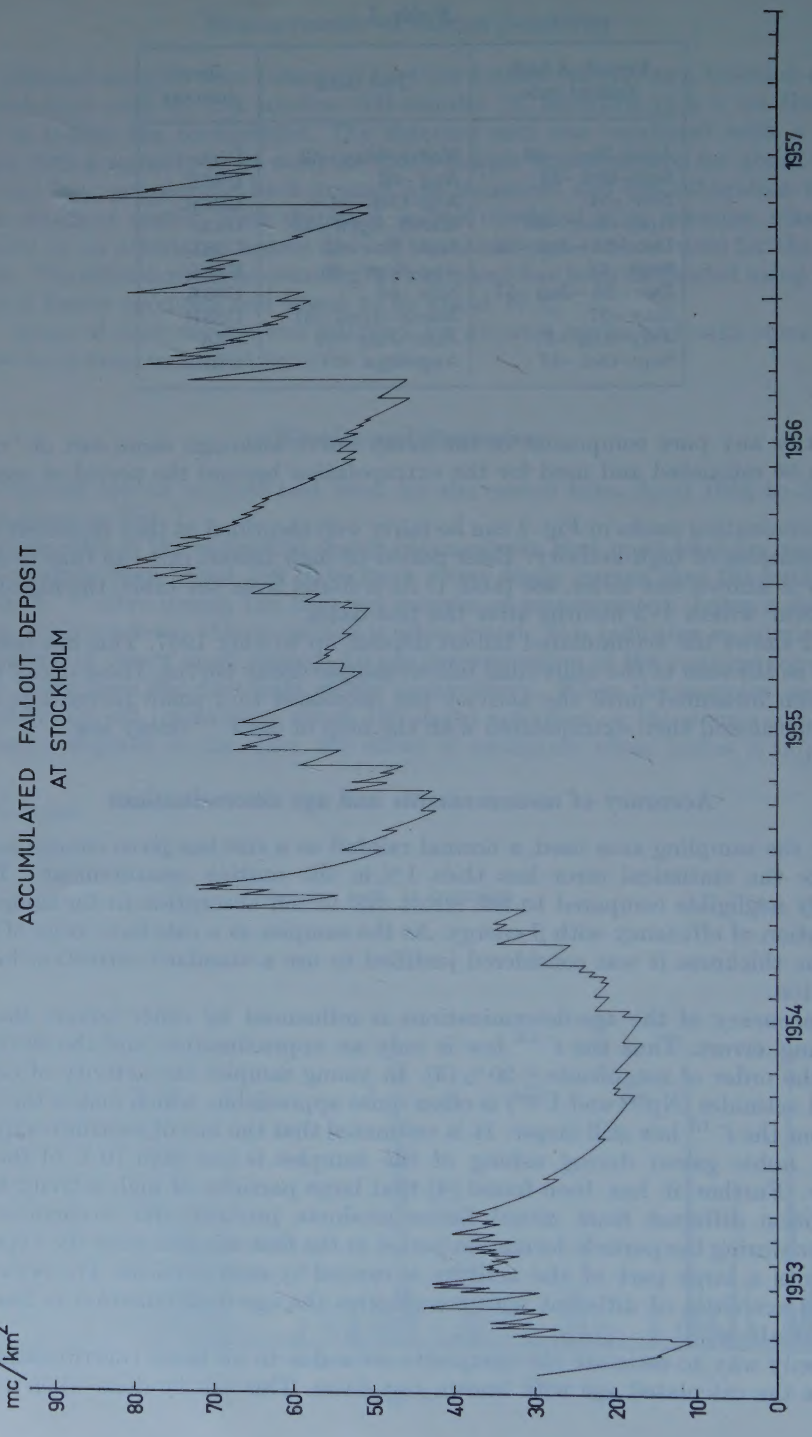
ACCUMULATED FALLOUT DEPOSIT
AT STOCKHOLM
mc/km²
90
80
70
60
50
40
30
20
10
0
1953
1954
1955
1956
1957

Fig. 2.

test has been carried out with a considerable intermission before and after. It is found in these cases that the age of the youngest component as a rule can be determined with an accuracy of about 10%.

Most of the difficulties in age-determinations mentioned above can be at least partly removed by determination of individual nuclides by means of γ-spectrometry or chemical separations instead of decay curve analysis. Determinations of this type have been made at this institute since 1955 and will be reported in a coming paper.

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